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EVALUATION OF POLLUTION CONTROL IN FOSSIL FUEL CONVERSION PROCESSES. GASIFICATION: SECTION 8. WINKLER PROCESS

C. E. Jahnig

Exxon Research and Engineering Company

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The report gives results of a review of the Winkler coal gasification process, from the standpoint of its potential for affecting the environment. The quantities of solid, liquid, and gaseous effluents have been estimated where possible, as well as the thermal efficiency of the process. For the purpose of reduced environmental impact, control systems, modifications, and alternatives which could facilitate pollution control or increase thermal efficiency are discussed, and new technology needs are pointed out.

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GASIFICATION: SECTION 8. WINKLER PROCESS

by

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TABLE OF CONVERSION UNITS

To Convert From	To	Multiply By
Btu	Calories kg:	0.25198
Btu/pound	Calories, kg./kilogram	0.55552
Cubic feet/day	Cubic meters/day	0.028317
Feet	Meters	0.30480
Gallons/minute	Cubic meters/minute	0.0037854
Inches	Centimeters	2.5400
Pounds	Kilograms	0.45359
Pounds/Btu	Kilograms/calorie, kg	1.8001
Pounds/hour	Kilograms/hour	0.45359
Pounds/square inch	Kilograms/square centimeter	0.070307
Tons	Metric tons	0.90719
Tons/day	Metric tons/day	0.90719

1. SUMMARY

The Winkler coal gasification process has been reviewed from the standpoint of its potential for affecting the environment. The quantities of solid, liquid and gaseous effluents have been estimated where possible, as well as the thermal efficiency of the process. For the purpose of reduced environmental impact, control systems, modifications, and alternatives which could facilitate pollution control or increase thermal efficiency are discussed, and technology needs are pointed out.

2. INTRODUCTION

Along with improved control of air and water pollution, the country is faced with urgent needs for energy sources. To improve the energy situation, intensive efforts are under way to upgrade coal, the most plentiful domestic fuel, to liquid and gaseous fuels which give less pollution. Other processes are intended to convert liquid fuels to gas. A few of the coal gasification processes are already commercially proven, and several others are being developed in large pilot plants. These programs are extensive and will cost millions of dollars, but this is warranted by the projected high cost for commercial gasification plants and the wide application expected in order to meet national needs. Coal conversion is faced with potential pollution problems that are common to coal-burning electric utility power plants in addition to pollution problems peculiar to the conversion process. It is thus important to examine the various conversion processes from the standpoint of pollution and thermal efficiencies and these should be compared with direct coal utilization when applicable. This type of examination is needed well before plans are initiated for commercial applications. Therefore, the Environmental Protection Agency arranged for such a study to be made by Exxon Research & Engineering Company under Contract No. EPA-68-02-0629, using all available non-proprietary information.

The present study under the contract involves preliminary design work to assure that conversion processes are free from pollution where pollution abatement techniques are available, to determine the overall efficiency of the processes and to point out areas where present technology or information is inadequate to assure that the processes are non-polluting.

All signficant input streams to the processes must be defined, as well as all effluents and their compositions. This requires complete mass and energy balances to define all gas, liquid, and solid streams. With this information, facilities for control of pollution can be examined and modified as required to meet environmental objectives. Thermal efficiency is also calculated, since it indicates the amount of waste heat that must be rejected to ambient air and water and is related to the total pollution caused by the production of a given quantity of clean fuel. Alternatively, it is a way of estimating the amount of raw fuel resources that are consumed in making the relatively pollution-free fuel. At this time of energy shortage this is an important consideration. Suggestions are included concerning technology gaps that exist for techniques to control pollution or conserve energy. Maximum use was made of the literature and information available from developers. Contacts were made with developers to up-date published information. Not included in this study are such areas as cost, economics, operability, etc. Coal mining and general offsite facilities are not within the scope of this study.

Other previous studies in this program to examine environmental aspects of fossil-fuel conversion processes covered various methods for

gasifying coal to make synthetic natural gas or low Btu gas. Reports have been issued on the Koppers, Synthane, Lurgi, CO_2 Acceptor, BIGAS, HYGAS, and U-Gas processes (1,2,3,4,5,6,7).

In the area of coal liquefaction, reports have been issued on the COED process of FMC (8) to make gas, tar, and char, as well as on the SRC process of Pittsburg & Midway Coal Mining Company to make a heavy liquid clean boiler fuel (9).

The present report presents our environmental evaluation of the Winkler process to gasify coal with steam and oxygen to make medium Btu gas. The study is based largely on literature references 10, 11, 12, 13, and 14 describing commercial plant operations. Acknowledgement is made to Mr. John M. Ferraro who made initial calculations to define the material balances for a Winkler gasifier.

3. SELECTION OF BASIS

During the period 1926-1960, a large number of commercial plants were built outside of the U.S. using the Winkler process to gasify coal. In most cases high purity oxygen is used rather than air, therefore this basis was selected for studying and evaluation. Although present units operate at about atmospheric pressure, designs at 6 atmospheres pressure are available and demonstration at higher pressure is planned. The present study is based on operating at 2 atmospheres.

A wide range of raw materials can be processed, including lignite, bituminous coal, anthracite, and heavy oil. However, to maximize carbon conversion, high reactivity is desirable, as is characteristic of lignites and younger coals. Our study is based on Leuna plant data for operation on a German brown coal (10), since the results may be pertinent to processing U.S. western coals. Operating conditions and oxygen consumption are based on this literature reference and are consistent with thermodynamic and heat balance calculation. The developer has since indicated that oxygen consumption may be decreased somewhat for new designs, together with a decrease in the amount of low level heat that must be rejected to air or water.

In order to define environmental aspects, scrubbing to remove sulfur was added, as well as a sulfur plant, oxygen plant, and other facilities needed to make the plant complete and self-sufficient. Plant size was set to provide net clean gas at the rate of 250×10^9 Btu/day, after supplying process requirements. The gas might be used as fuel or reducing gas, or it could be converted to ammonia, chemicals, SNG, or oil.

4. PROCESS DESCRIPTION

Lignite type coal is gasified at about 1700°F and 2 atmospheres in a turbulent bed of particles using oxygen and steam, to make medium Btu gas for fuel or synthesis. Some of the residual char is withdrawn from the bottom of the gasification reactor, but most of it is blown overhead as a result of the high gas velocity of 5-10 ft/sec. Most of the entrained char is collected in cyclones for disposal, and the gas is then cooled and cleaned up to remove residual dust and sulfur.

An overall flowplan of the process is shown in Figure 1. The process can be subdivided into a sequence of steps, each of which will be described in the following sub-sections: (1) coal preparation, (2) gasification, (3) cooling and scrubbing, (4) sulfur removal, and (5) auxiliary facilities.

4.1 Coal Preparation

This section of the plant includes storage and handling, drying, and crushing. It is assumed that coal cleaning is not required, or that it is carried out elsewhere. Storage requirements will depend upon the specific situation but may provide for example 30 days reserve.

Drying may not always be needed, since it is only necessary to avoid surface moisture which would cause problems in handling and crushing. Rotating tray dryers are used, and for this study a moisture removal of 5% on feed has been taken. Cool stack gas is recycled to control gas inlet temperature so as not to drive off volatiles. Stack temperature is 350-400°F, resulting in good fuel efficiency. Coal can be used as fuel if flue gas desulfurization is provided, but instead of this we have used part of the clean product gas as fuel to the dryer, with bag filters on the vent gas to control dust emissions. Coal is crushed to 0-8mm, and sent to the gasifier feed hopper.

4.2 Gasification

Coal from the feed hopper is fed to the gasifier by means of screw feeders which give the necessary pressure seal. As shown in Figure 2, steam and oxygen are added near the bottom of the reactor, maintaining the particles in a turbulent bed where reaction takes place without reaching temperatures that would fuse the ash. Typically, the bed may be at about 1700°F so that tar and heavy hydrocarbons are destroyed by gasification reactions.

Considerable fines are entrained from the bed, consequently supplemental oxygen and steam are added just above the bed to help consume them. Heat exchange surface in the dilute phase above the bed removes heat to protect refractories and for temperature control, generating useful steam. The raw gas is cooled to about 1300°F before the gas leaves the reactor, in order to prevent fused deposits in the downstream waste heat boiler. Condensate can also be injected into the gas for temperature control and also provides backup or emergency cooling.

With high reactivity coal, conversion of carbon in the coal feed may be about 90%. The unconverted carbon is in the char by-product, and

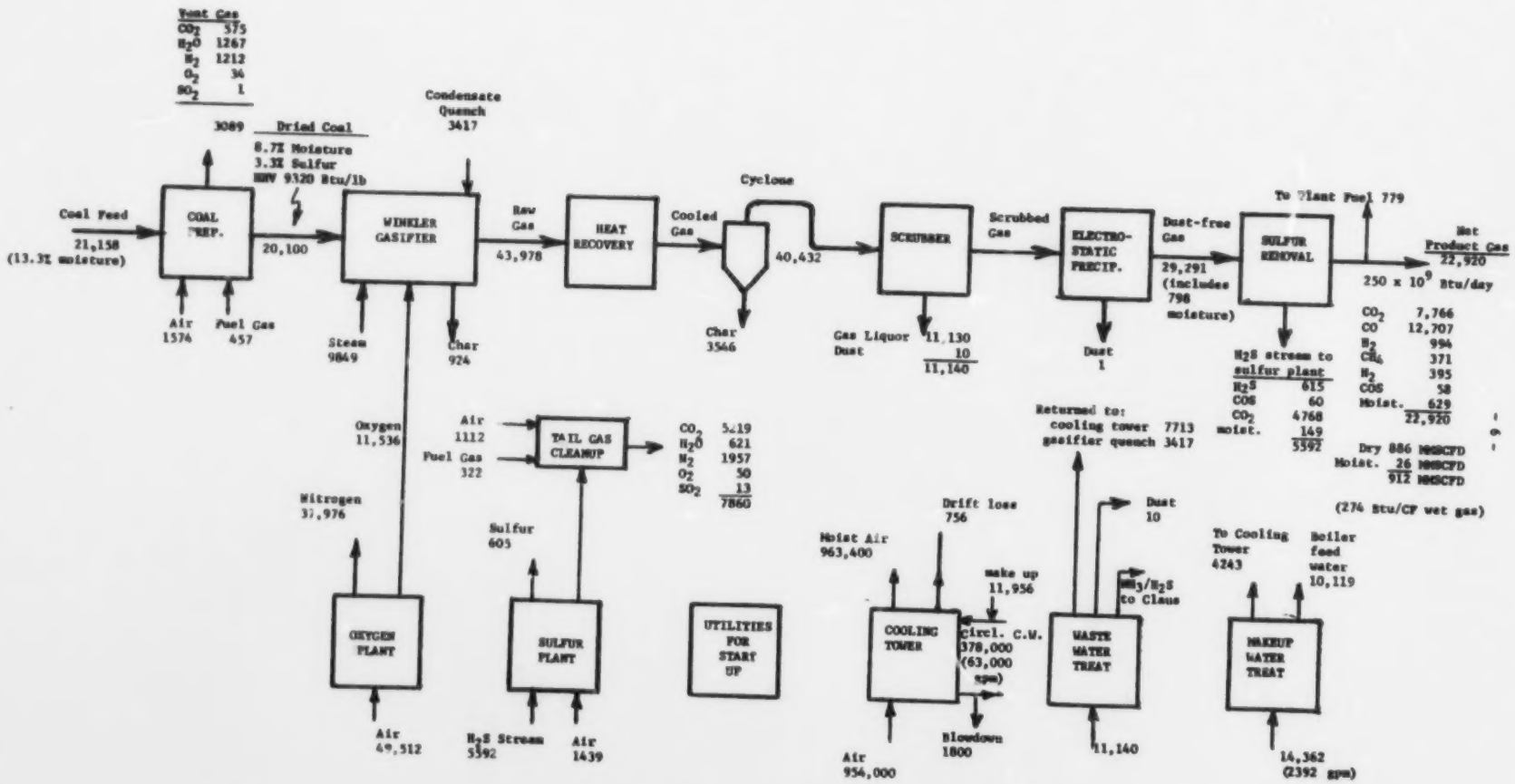
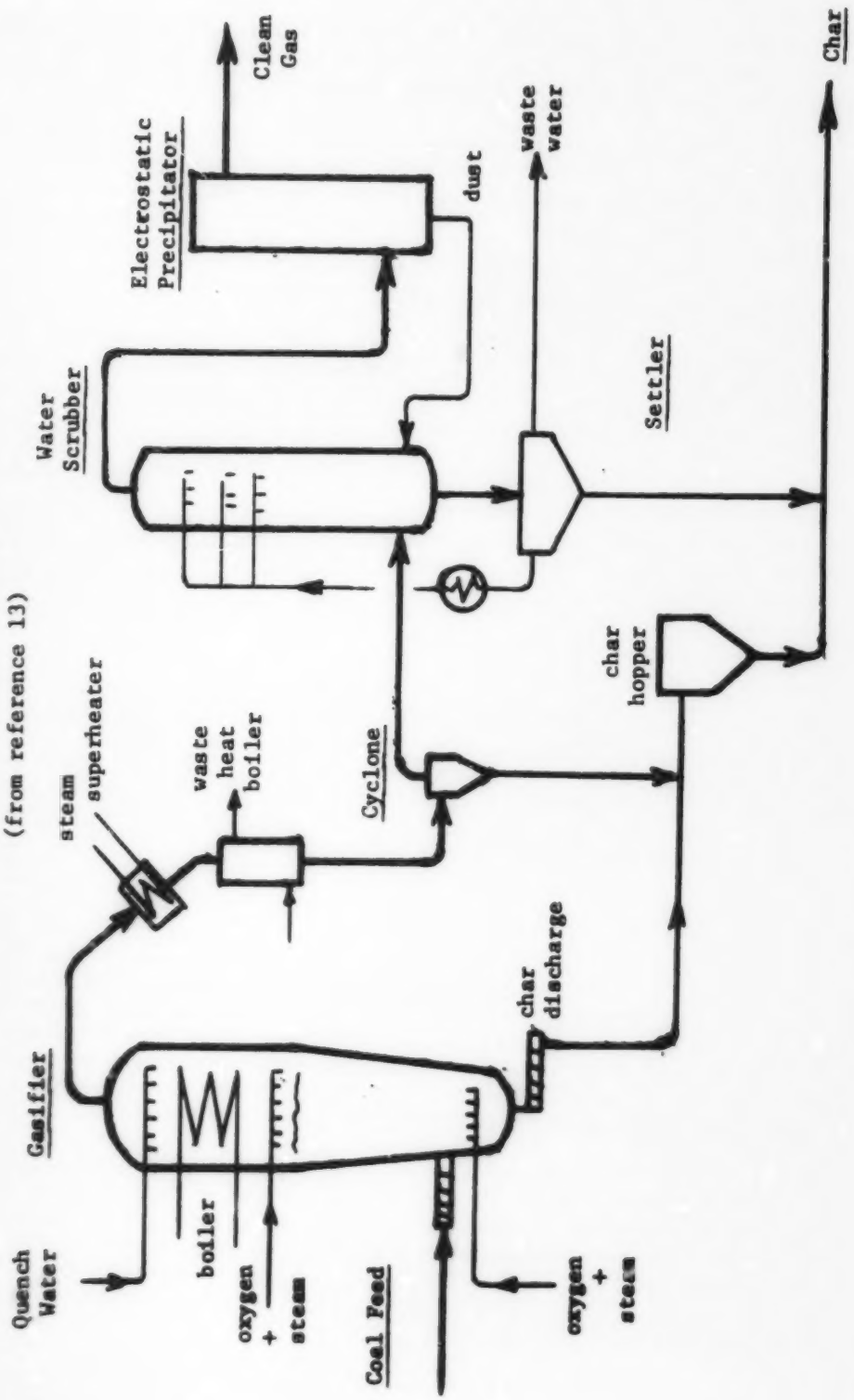


FIGURE 1
Flowplan for Winkler Process with Auxiliary Facilities
Numbers are flowrates in tons/day except as noted.

FIGURE 2

WINKLER GASIFICATION SYSTEM

(from reference 13)



represents a significant loss of heating value unless it is used. Part of the rejected char is withdrawn from the bottom of the gasifier, and the remainder is recovered by a cyclone separator on the exit gases.

Steam fed to the gasifier amounts to about 0.5 pound per pound of coal feed, while steam conversion including moisture in the coal feed is 27%. Oxygen consumed is 0.57 pounds per pound of coal feed for this specific design basis that does not use preheating on the streams fed to gasification.

4.3 Gas Cooling and Dust Removal

Hot raw gas leaving the reactor at about 1300°F passes through an exchanger to superheat steam, followed by a waste heat boiler and a cyclone to remove entrained char. The gas then goes to a scrubbing tower where it is cooled by direct contact with recirculated water.

Most of the particulates are removed by scrubbing and are separated from the water in a settler. They are included with the char for disposal. Clarified water is cooled by indirect exchange with cooling water before it is recirculated to the scrubber. Net production of this water or gas liquor constitutes sour water containing H_2S, ammonia, cyanides, etc., present in the raw gas. The sour water is processed in waste water treating so that it can be reused.

Since the scrubbed gas will still contain a small amount of dust, it is passed through an electrostatic precipitator for final cleanup. It can then be compressed, further processed, or used as desired. Traces of containinants may remain in the gas after scrubbing, such as ammonia, sulfur, oil, etc., especially during upsets or start up. Depending on the intended use, further cleanup may be necessary. In some applications the electrostatic precipitation may not be needed.

4.4 Sulfur Removal

The next processing step on the gas is sulfur removal by scrubbing with a suitable solution, such as amine, hot carbonate, or a glycol type solvent. These can be regenerated by stripping to give a concentrated H_2S stream that is sent to sulfur recovery. For this study scrubbing with hot carbonate is assumed, since it will remove perhaps half of the carbonyl sulfide present in the gas, and some 10% of the total sulfur will be in this form which is not removed effectively by amines.

As an alternative, H_2S in the gas might be converted directly to free sulfur by using an absorption/oxidation type process such as is offered by Stretford, Takahax, or IFP. In effect, this route would combine the sulfur recovery plant with scrubbing to remove H_2S. Sulfur compounds other than H_2S are not usually removed by such systems.

4.5 Auxiliary Facilities

In order to make a realistic and thorough evaluation of environmental impacts, a complete and self-sufficient plant must be considered, including items such as oxygen plant, sulfur recovery, water treating, and

utilities generation. Oxygen is supplied from a conventional air liquefaction plant. The amount is large, equal to 11,536 tons/day. For sulfur recovery, a Claus plant is included with tail gas cleanup using one of the many processes offered for this service. Details and alternatives are discussed more fully in previous reports of this series. Gas sent to the Claus plant from acid gas treatment contains about 15 vol. % sulfur compounds (mainly H_2S) and 85 vol. % CO_2, on a dry basis. A small amount of clean product gas is used as fuel to incinerate tail gas on the sulfur plant.

A major item is waste water treating on the gas liquor condensed in the scrubber. Flow rate is 11,140 tons/day, and cleanup is required to remove particulates, contaminants such as compounds containing sulfur, nitrogen, or oxygen, as well as arsenic, cadmimum, lead, chlorine, fluorine, and other trace elements that are known to be volatile at conditions in the gasifier. This water stream must be thoroughly cleaned up in any case, and then represents a very desirable makeup water for the plant. Facilities include sour water stripping, biological oxidation (biox), and sand filtration prior to using it as cooling tower makeup. Production of phenols is expected to be relatively low at the conditions used in the gasifier (1700°F) so that solvent extraction to remove large amounts of phenols is not included. Definitive information should be obtained on the nature of the gas liquor resulting from the Winkler operation.

Other auxiliary facilities include treatment of makeup water for the cooling water system and for boiler feed water, plus plant utilities such as steam and electric power. It appears from the balances that the plant should be self-sufficient in steam and power during normal operation, although provision must also be made for startup. As far as energy balances and thermal efficiency are concerned, no coal or clean product gas need be consumed to generate plant utilities.

The cooling tower has a very important potential environmental impact in that the air flow through it is by far the largest stream in the whole plant. Any potential contamination of the air is a major concern, such as may result from leaks that could contaminate the circulating cooling water. Moreover, evaporation in the cooling tower is the primary factor determining net water makeup required by the process.

5. PROCESS STREAMS and EMISSIONS

A block diagram is given in Figure 3 indicating the various streams for the plant, with a description of these in Table 1. Process streams are shown as well as those streams actually released to the environment. The latter are indicated in Figure 3 by heavy dashed lines and in Table 1 by asterisks, while the other streams are returned to the process. Environmental aspects and control techniques will now be discussed for the various gas, liquid, and solid streams, in the order of processing steps as indicated in the preceding section on process description.

5.1 Coal Preparation

A first consideration is the handling and storage of large amounts of coal feed. Delivered coal must be loaded on conveyors, with transfer to and from storage piles. Such operations necessarily tend to create problems due to noise, dust nuisance, and spills. These facilities should be enclosed as much as possible, with plans and equipment provided for cleanup. A dust collector system is desirable, operating at below atmospheric pressure to collect vent gas and pass it through bag filters.

Storage piles are an additional concern since wind can disperse fine particles. In some cases consideration has been given to covering the coal pile, or coating it, for example with heavy tar. The pile is very large, over 600,000 tons for 30 days storage, requiring an area of about 10 acres. Coal piles are also liable to spontaneous combustion, calling for special attention and plans for control, together with provision for extinguishing fires if they occur (15). The obnoxious fumes, sulfur, and odor from this type of fire is well known. Previous reports in this series include further discussion of the general subject (e.g. 5) but for any specific project, a very careful and thorough evaluation and definition of facilities is needed.

Noise control should be carefully considered since it is often a serious problem in solids handling and size reduction. If the crushing equipment is within a building, the process area may be shielded from undue noise but additional precautions are needed from the standpoint of personnel inside the building. Other sources of noise include compressors or other rotating equipment, furnaces, vents, valves, flares, etc.

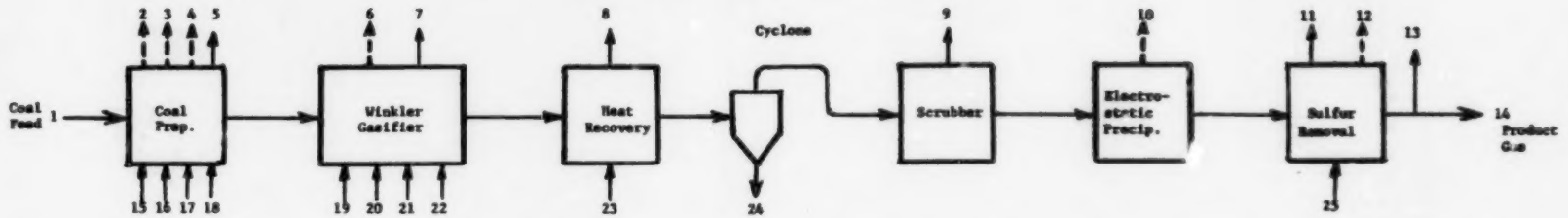
The present design is based on processing run of mine lignite. If the process were used on bituminous coal then some cleaning or washing operation would normally be used. It should be pointed out that coal cleaning and washing results in rejection of a large amount of refuse and fines, often 25% of the mined coal, with major environmental impacts as discussed in previous reports in this series.

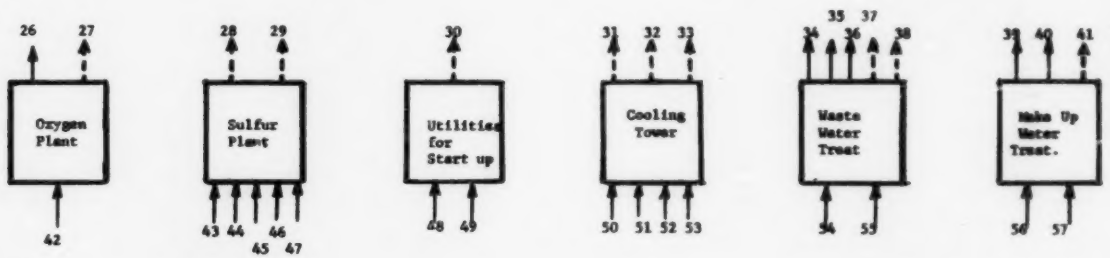
Coal is crushed through 4 mesh and fed to a dryer where surface moisture is removed. The dryer is designed to avoid overheating coal particles, which would release volatiles. To maximize fuel efficiency, combustion is carried out with only 10% excess air, and dryer offgas is recycled to temper the hot gas to about 700°F before it enters the dryer.

FIGURE 3

WINKLER GASIFICATION PROCESS

Plant Streams and Effluents
(See Table 1 for details on numbered streams)





Note: Streams actually released to the environment are shown by heavy dashed lines, other streams are returned to process.

TABLE 1

WINKLER GASIFICATION PROCESS
PLANT STREAMS AND EFFLUENTS (see Figure 3)

Stream No.	Identification	Flow, tons/day	Comments
1	Coal feed	21,158	Cleaned coal feed with 13.3% moisture (see Table 5 for specifications)
*2	Wind	---	Action of wind on storage pile may cause dusting or fires.
*3	Rain	e.g. 6" in 24 hr.	Rain action on storage pile can wash out fines, cause leaching of sulfur, metals, and organics--similar to acid mine water, should be collected and sent to pond for use as make up.
*4	Vent Gas	3089	Flue gases from coal dryer--see Figure 1 for composition.
5	Dust	---	Recovered from vent gas on coal dryer and included in feed to gasifier.
*6	Char	924	Withdrawn from bottom of gasifier. Contains 42% carbon and should be burned using environmental controls so that heating value is recovered.
7	Steam	9045	High pressure steam (600 psig) generated in gasification section. (see Table 7).
8	Steam	6030	125 psig generated from waste heat in raw gas (see Table 7).

Table 1 (con't.)

Stream No.	Identification	Flow, tons/day	Comments
9	Gas Liquor	11,140	Water condensed from scrubbing raw gas--contains ammonia, sulfur compounds, and dust, etc., and is sent to waste water treating to clean up for reuse.
*10	Dust	1	Minor amount of dust removed by electrostatic precipitator to make clean product gas.
11	H_2S Stream	5592	Sulfur compounds together with CO_2 from sulfur removal on gas--sent to sulfur plant. See Figure 1 for composition.
*12	Chemical Purge	---	Some of chemical scrubbing solution used in sulfur removal is lost or purged to maintain capacity and constitutes an effluent from the plant.
13	Plant Fuel Gas	779	Part of clean product gas is used as fuel in coal dryer and Claus tail gas incinerator.
14	Product Gas	22,920	Net clean product gas. See Table 6 for details.
15	Wind	---	Wind action on Storage pile.
16	Rain	e.g. 6" in 24 hrs.	Rain onto storage pile.
17	Fuel Gas	457	Part of product gas used as fuel in coal dryer.
18	Air	1574	Air for combustion of fuel gas in coal dryer.

Table 1 (con't.)

Stream No.	Identification	Flow, tons/day	Comments
19	Steam	9849	Steam added to gasifier.
20	Oxygen	11,536	Oxygen added to gasifier.
21	Boiler feed water	9045	To generate steam on gasifier, See item 7.
22	Quench Water	3417	Treated sour water--added at outlet of gasifier to temper gas and prevent slag deposits on waste heat boiler.
23	Boiler Feed Water	6030	To generate steam in waste heat boiler after gasifier. See item 8.
24	Char	3546	Residue left after gasification and entrained with raw gas.
25	Chemical Makeup	---	Chemicals are used in sulfur removal (e.g. amine, or carbonate) and are lost or purged so that a corresponding chemical makeup is required.
26	Oxygen	11,536	Produced in oxygen plant and sent to gasifier.
*27	Nitrogen	37,976	By product from oxygen production and vented to air. Should be clean.
*28	Sulfur	605	By product recovered in sulfur plant, to be sold.
*29	Tail Gas	7860	From tail gas cleanup after Claus sulfur recovery plant. See Figure 1 for composition.
*30	Flue Gas	---	From utility boiler. Not used during normal operation but is needed for startup. Low sulfur oil fuel may be used to avoid pollution problems at startup.

Table 1 (con't.)

Stream No.	Identification	Flow, tons/day	Comments
*31	Air	963,400	Moist air from cooling tower--contains 9400 tons/day of evaporated water.
*32	Mist	756	Nominal drift loss of cooling water lost by entrainment in air.
*33	Blowdown	1800	Purge from cooling water circuit to control buildup of dissolved solids--will contain cooling water additives such as chromate and chlorine so may require treatment before disposal.
34	Quench Water	3417	Treated waste water used as quench at gasifier outlet. See item 22.
35	Makeup Water	7713	Treated waste water used as makeup on cooling water.
36	Sour Gas	---	H_2S, NH_3, etc. stripped from sour water and sent to Claus plant for incineration and disposal.
*37	Dust	10	Nominal amount of dust in sour water from scrubbing which is recovered in settler and can be included with char for disposal.
*38	Sludge	---	Sludge produced in biological oxidation which may be buried or incinerated.
39	Makeup Water	4243	Fresh water makeup needed to balance cooling water circuit.
40	Makeup Water	10,119	Net boiler feed water makeup required after crediting condensate that can be collected and reused.

Table 1 (cont'd.)

Stream No.	Identification	Flow, tons/day	Comments
*41	Sludge	---	From chemical treating of makeup water, e.g. lime sludge. See Table 10.
42	Air	49,512	Air processed in oxygen plant.
43	H_2S Stream	5592	Sent to Claus unit from sulfur removal section.
44	Sour Gas	---	From sour water stripping--sent to Claus unit for incineration and disposal.
45	Air	1439	Air for incineration in Claus unit.
46	Fuel Gas	322	Part of clean product gas used to incinerate tail gas from Claus unit prior to tail gas cleanup.
47	Air	1112	Used to burn fuel in item 46.
48	Fuel Oil	---	Low sulfur fuel oil used for plant startup. Not needed during normal operation.
49	Air	---	Combustion air for item 48.
50	Air	954,000	Air flow into cooling tower.
51	Cooling Water	378,000	Circulating cooling water.
52	Makeup Water	11,956	Makeup water to cooling water circuit—the sum of items 35 and 39.
53	Additives	---	Chemicals added to cooling water system to control corrosion (chromates) and fouling (chlorine) etc.
54	Gas Liquor	11,140	Foul water from scrubber fed to waste water treating.
55	Chemicals	See Table 10	Chemicals used to treat waste water, such as lime for pH control and to precipitate fluorides. Nutrients may be needed in biox unit.

Table 1 (cont'd.)

Stream No.	Identification	Flow, tons/day	Comments
56	Makeup Water	14,362	Total makeup water to plant. See Table 9.
57	Chemicals	See Table 10	Chemicals used to treat makeup water, such as lime, alum, acid, caustic, etc.

* These streams are actually released to the environment, other streams are returned to the process.

Low excess air also decreases the volume of vent gas compared to some other drying systems that may use as much as 100% excess air in order to facilitate drying.

To prevent sulfur emission in the dryer vent gas, part of the clean product gas is used for fuel, rather than burning coal. This consumes 2% of the product gas. Dust control is also needed, therefore bag filters are provided, with the fines being returned to the gasifier. As extensive drying is not essential for process operability, consideration can be given to ommiting the dryer and allowing for increased heat load on the gasifier.

5.2 Gasification

Coal is fed to the gasifier from a feed hopper, using screw conveyors or feeders. As this system is enclosed, dust and gas can be contained to prevent emissions to the environment. Attention should be given to potential leaks, operating procedures, and maintenance, to assure that this is the case. Gas from purging and blanketing must be collected, and can be sent to bag filters, for example on the coal dryer.

The major effluent from the gasification section is char that is withdrawn from the bottom of the reactor. Screw conveyors transfer the char to enclosed storage hoppers, from which it is withdrawn from disposal. While this portion of the char consists of coarser particles due to elutriation in the gasifier, there can still be a dusting problem associated with handling and disposal. Dusting can be controlled by proper planning and design, possibly using water sprays and partial wetting of the char. Inadvertent spills of char can also be a problem, so consideration of this is needed with provision for cleaning up spills if they occur. The same applies generally to solids handling operations, such as coal storage, preparations and feeding.

Based on the literature reference used as a bases (10), rejected char from the gasifier contains about 40% carbon, therefore it will be desirable to consider ways to recover the heating value it represents. One possibility is to burn it in a furnace, but environmental controls would be needed to give acceptable sulfur and dust emissions. Flue gas scrubbing would be one method for control. A second and much larger stream of char is rejected from the gas cleaning section of the plant, which also has a high carbon content. Aspects of char disposal will be discussed further in the following section 5.3 relating to gas cleanup.

5.3 Gas Cooling and Dust Removal

A waste heat boiler recovers useful heat from the raw gas leaving the gasifier. Steam superheating is also provided, and all plant steam and power requirements can be supplied using by product steam from the process. Considerable char is entrained from the gasifier and passes through the heat recovery exchangers before being collected in cyclone separators. The collected char is relatively fine and contains a substantial amount of carbon, roughly 30% for this study case. It is removed from the system to a storage hopper for ultimate disposal.

The char streams from a Winkler plant might be used as land fill, although the resulting loss in carbon would represent 11.5% of the

heating value in the coal feed. One possibility is to burn the combined char streams left after gasification, as is done in the commercial Winkler plant at Kutahya, Turkey (16) where the char is burned in a steam boiler. This same approach could be used in the United States except that stack cleanup would be required in order to control emissions of dust and sulfur. Spent char might also be used as fuel in cement manufacture, or it could be considered and evaluated as an adsorbent for use in water treating.

The next step in the gas cleanup sequence is water scrubbing to give additional dust removal and at the same time cool the gas. Water is condensed from the gas, giving a gas liquor containing many contaminants present in the raw gas, including ammonia, H_2S, and probably small amounts of phenols, cyanides, hydrocarbons, etc., and dust. In addition, it is known that certain trace elements are at least partially volatile at gasification conditions; consequently, they may be present in the raw gas and have to be removed. Some condensation and buildup of volatile materials on entrained char or dust can be expected and the potential environmental impacts need to be defined. Many of the volatile trace elements are very toxic, such as; arsenic, cadmium, lead, and fluorine. The subject of trace elements calls for special attention and is discussed in a separate section. The gas liquor is not released directly to the environment, but goes to waste water treating, and will be discussed in Section 5.5 on auxiliary facilities.

In some applications additional dust removal may be needed to prevent plugging of catalyst beds or to protect equipment such as compressors, therefore an electrostatic precipitator is provided in the study case. The small amount of dust recovered in it can be included with the rejected char for disposal. In some situations the electrostatic precipitator may not be needed, for example, dust removal might be achieved in the subsequent sulfur removal operation which usually will involve efficient scrubbing with liquid.

5.4 Sulfur Removal

There are a number of alternative processes that could be used to recover H_2S from the gas such as scrubbing with amine or modified amines, hot carbonate, glycol type solvent, or refrigerated methanol. Carbonyl sulfide is also present in the gas, equivalent to perhaps 10% of the total sulfur, and should be removed. Although conventional amine scrubbing is not effective for removing carbonyl sulfide, part or most of it can be taken out by scrubbing with hot carbonate, glycol, or refrigerated methanol. Our study assumes that hot carbonate or glycol scrubbing will be used, giving COS removal with moderate utilities consumption. It may be desirable to include a hydrolysis step to convert COS to H_2S plus CO_2 prior to scrubbing for acid gas removal.

The H_2S stream is sent to a Claus type sulfur plant with tail gas clenaup. No specific attempt is made to remove CO_2 from the gas, assuming that the primary need is to remove sulfur. However, considerable CO_2 is removed along with the H_2S, such that the stream to sulfur recovery contains about 15% H_2S and 85% CO_2, on a dry basis.

A possible arrangement to consider for sulfur removal would combine H_2S removal with conversion to by product sulfur in one operation, using an absorption/oxidation type process. Such processes are offered by Stretford, Takahax, and IFP. They use a catalytic scrubbing solution to absorb H_2S, which is then oxidized to free sulfur using combined oxygen which is held by the solution. In effect, the absorption of H_2S and its conversion to free sulfur are combined into a single operation. An advantage of this route is that very efficient removal of H_2S is practical at low pressure. Also, CO_2 is not removed, which may or may not be an advantage. A disadvantage is that these processes are not usually effective for removing other forms of sulfur such as carbonyl sulfide; however, it may be possible to hydrolyze these other sulfur compounds to H_2S prior to sulfur removal by incorporating a bed of alumina or bauxite catalyst in the gas cooling system at an appropriate point to give the proper temperature of 500-700°F. (17).

In general, the scrubbing solutions used for sulfur removal will degrade due to side reactions or accumulation of inert materials. A small amount of solution is usually purged to maintain capacity or activity. This constitutes a chemical effluent from the plant that must be disposed of. To the extent that it is combustible, incineration may offer a means of disposal, but for materials such as potassium carbonate or metals such as vanadium, other methods of disposal will have to be defined.

5.5 Auxiliary Facilities

These include the oxygen and sulfur plants, plus utilities supply and water treating. The oxygen plant is a large consumer of utilities, but has no objectionable effluents. The waste nitrogen stream is clean, and the only other effluent is some water condensed from the air, which can be used as boiler feed water.

In addition to byproduct sulfur, the sulfur plant releases treated tail gas which is comparable to flue gas from combustion of low sulfur fuel. A typical sulfur recovery is 99% for a Claus plant with tail gas cleanup, giving about 1600 wt. ppm of sulfur dioxide in the stack gas emitted to the atmosphere. This would be comparable to the flue gas from burning a char of about 1.0% sulfur. Some clean product gas is burned with air to provide incineration required for tail gas cleanup.

In some cases tail gas cleanup is carried out by reducing sulfur compounds in the Claus plant tail gas to H_2S, which is then removed by scrubbing, for example with amine. In other cases the tail gas may be incinerated to form SO_2 which is then scrubbed out. From an environmental control standpoint, either approach should be satisfactory and the choice may reflect other considerations. Chemical solutions are normally used for scrubbing in tail gas cleanup, and undergo some degradation such that a small amount must be purged. Disposal of this purge solution can be handled as discussed in the preceding section 5.4 on sulfur removal.

The sulfur plant will, of course, be a likely source of odors, which must be carefully controlled. Suitable designs and operating techniques have been established for clean operation of sulfur plants, and for handling, storing, and shipping sulfur.

Other auxiliary facilities include supply and distribution of steam and electric power. As mentioned, these can be supplied as by-products from waste heat recovered in the process, so that no utility boiler is needed during normal operation. Provision will be needed for startup, etc. Potential pollution from furnace flue gas on the utility boiler is, therefore, not a problem, nor is it necessary to consider burning part of the clean product gas in order to supply utilities. For startup conditions it would be reasonable to depend on storage of low sulfur oil, rather than use coal which would require additional pollution control facilities.

A moderate size cooling tower is required to supply cooling water used in the process. It has by far the largest emission from the plant, namely 954,000 tons/day of air plus 9,400 tons/day of evaporated water. As discussed in previous reports, it is imperative to keep contaminants out of the cooling water circuit, so that they can not then be stripped out into the air passing through the cooling tower. There are also the usual questions of drift loss and potential plume or fog formation which must be considered and evaluated. Proper design and placement of the cooling tower can aleviate or avoid potential problems such as effect on public highways.

A further effluent from the cooling water circuit is blowdown of purge water to control buildup of dissolved solids in the cooling water. Additives used in the cooling water circuit will necessarily appear in the blowdown stream, together with dissolved solids that accumulate and buildup. Chlorine is often added to cooling water to inhibit algae growth and the fouling of heat exchanger surfaces, while chromates or other chemicals are usually added to combat corrosion. These additives will then be in the blowdown water, which may also include products of corrosion such as copper, etc. from extensive heat transfer surfaces.

As is usually the case, the only point where soluble salts can leave the plant is in the cooling tower blowdown. Thus, dissolved solids in the plant makeup water, such as sodium sulfate and chloride, become concentrated due to evaporation of water in the cooling tower. If the makeup water contains 500 ppm of such salts, they will then buildup to 2500 ppm in the blowdown water for the purge rate used in this evaluation. Such water would be considered brackish, and unsuitable even for irrigation, and at inland locations may present a disposal problem. In one proposed plant it is sent to an evaporation pond, where the dried salts are stored. It would be desirable to have better ways of handling the blowdown water, for example recovering the water content for reuse in an indirect evaporator using waste heat.

Additional auxiliary facilities provide treatment of waste water and plant makeup water. The rates are shown in Figure 1 and Table 1. Waste water cleanup will include sour water stripping to remove ammonia and H_2S. The ammonia may be sufficient to warrant recovery, but the small amount of H_2S dissolved at this low pressure can be sent to the sulfur plant for disposal. The amount of phenols, HCN, and oil or other hydrocarbons is expected to be minor at the gasification conditions of 1700°F. and low pressure. These can probably be removed adequately by biological oxidation (biox) with 7-10 days retention time, before the sour water is used as cooling tower makeup. It may be necessary to also use filtration and treatment with activated carbon to clean up the sour water. In fact, the spent char may be useful for this purpose. Effluents to the environment from waste water treating are: byproduct ammonia, ash and solids removed by the settler, oil or other contaminants removed during cleanup, together with sludge from the biox unit. If chemical treatments are used, such as lime, these will also contribute effluents. In addition, there will be trace elements that vaporize in the gasifier and accumulate in the sour water. These must be removed and recovered as byproducts, or deactivated for disposal in a safe and satisfactory manner. The subject will be discussed in more detail in Section 8 on Trace Elements.

Finally, facilities are needed to treat the makeup water needed by the plant. This usually includes treatment with lime, alum, etc., as well as demineralization to prepare boiler feed water. The latter may use water softeners, and ion exchange resins that are regenerated by back washing with acid or caustic. Obviously, all chemicals used and consumed in treating will appear in plant effluents at some point, together with materials removed from the makeup water. Further definition is needed for each specific case, but the sludge from water treating can probably be disposed of along with the char, or separately as land fill.

6. SULFUR BALANCE

Nearly all of the sulfur in the coal appears in the raw gas leaving the gasifier, from which it can be separated and sent to a Claus plant for sulfur recovery. The latter gives 99% sulfur recovery with tail gas cleanup. Of the total sulfur in the raw gas, 10% of it may be in the form of carbonyl sulfide plus small amounts of CS_2 and other sulfur compounds, half of which is recovered and sent to the sulfur plant. For this particular study, byproduct sulfur accounts for 91.2% of the sulfur entering with the coal feed. An overall sulfur balance is shown in Table 2.

In calculating sulfur content of the product gas, it was assumed that half of the carbonyl sulfide in the raw gas would be removed and sent to sulfur recovery, while the remainder would appear in the product gas. This could change depending on the technique used for gas cleanup, and it would be desirable to have methods giving more complete sulfur removal from the gas with low energy consumption.

The rejected char may possibly have a relatively low sulfur content compared to the feed coal, such that it might be burned without requiring special provision to decrease sulfur emission. Detailed plant data to confirm this are not readily available in the literature but should be examined where possible. Other background (8,18) suggests that gasification conditions may tend to desulfurize the char sufficiently so that the byproduct char might be marketed as a low sulfur solid fuel, at least in some cases. If this is true, it could turn the problem of char disposal into a potential advantage. In effect there would be a credit for desulfurizing part of the coal feed, and there would be less incentive to operate at high carbon conversion in the gasifier.

TABLE 2

SULFUR BALANCE--WINKLER PROCESS

	tons/day	%
Sulfur in coal feed	663	100
Sulfur in net product gas	31	4.7
Sulfur in plant fuel gas	1	0.2
By product sulfur from Claus plant	605	91.2
Sulfur in tail gas of sulfur plant	6	0.9
Sulfur in char and ash (est.)	20	3.0
	663	100.0

7. THERMAL EFFICIENCY

Heating value of the net clean product gas from the process is 66.8% of that for the coal consumed as shown in Table 3. This is for the complete plant including auxiliaries such as oxygen plant, sulfur plant, and utilities. It does not include any credit for the char byproduct, which would bring the total heating value of products to 78.3% of that for the coal feed. Clearly there is a large incentive to recover the heating value contained in the char. If it is low enough in sulfur, it can be burned as fuel using proper dust recovery. If the char is high in sulfur, the emphasis should be placed on efficient gasification to minimize the residual carbon content of the char.

Distribution of losses that decrease thermal efficiency are shown in Table 3. Most of the loss is rejected to cooling water or in air coolers, representing low level heat that is impractical to recover and use with present conventional technology.

Thermal efficiency will of course depend upon the specific coal used, particularly the ash and moisture content, and the coal reactivity which affects carbon level in the rejected char.

TABLE 3

THERMAL EFFICIENCY--WINKLER PROCESS

	10^9 Btu/day	%
Coal feed @ 9320 Btu/lb HHV	374	100
Net product gas (wet) 912 MM SCFD @ 274 $\frac{Btu}{CF}$	250	66.8
Losses:		
Fuel gas to coal dryer	5	1.3
Fuel gas to Claus incinerator	4	1.1
Carbon in withdrawn char	43	11.5
In H_2S to sulfur recovery	10	2.7
Power consumers	5	1.3
To air cooling	18	4.8
To cooling water*	27	7.2
Heat losses and miscellaneous	12	3.3
	124	33.2

* Approximately 20 x 10^9 Btu/day goes to evaporate water, and the rest goes to sensible heat of the air flowing through the cooling tower.

8. TRACE ELEMENTS

Coal contains many trace elements present in less than 1% concentration that need to be carefully considered from the standpoint of potential impact on the environment. Many of these may volatilize to a small or large extent during processing, and many of the volatile components can be highly toxic. This is especially true for mercury, selenium, arsenic, molybdenum, lead, cadmium, beryllium and fluorine. The fate of trace elements in coal conversion operations, such as gasification or liquefaction, can be very different than experienced in conventional coal fired furnaces. One reason is that the conversion operations take place in a reducing atmosphere, whereas in combustion the conditions are always oxidizing. This maintains the trace elements in an oxidized condition such that they may have more tendency to combine or dissolve in the major ash components such as silica and alumina. On the other hand, the reducing atmosphere present in coal conversion may form compounds such as hydrides, carbonyls or sulfides which may be more volatile. Studies on coal fired furnaces have indicated that smaller particles in fly ash contain a higher concentration of trace elements, presumably due to volatilization of these elements in the combustion zone and their subsequent condensation and collection on the fly ash particles (19). Other studies on coal fired furnaces are pertinent (20,21,22) and some of these report mass balances on trace elements around the furnaces (23).

Considerable information is available on the analyses of coal, including trace constituents, and these data have been assembled and evaluated (24,25,26). A few experimental studies have been made to determine what happens to various trace elements during gasification (27,28). As expected, these show a very appreciable amount of volatilization on certain elements. As an order of magnitude, in this specific Winkler design, each 10 ppm of element volatilized would amount to about 400 pounds per day.

In order to make the picture on trace metals more meaningful, the approximate degree of volatilization shown for various elements has been combined with their corresponding concentration in a hypothetical coal (as typical), giving an estimate of the pounds per day of each element that might be carried out with the hot gases leaving the gasifier. Results are shown in Table 4 in the order of decreasing volatility. Looking at the estimated amounts that may be carried overhead, it becomes immediately apparent that there can be a very real problem. For each element the net amount carried out in the gas leaving the gasifier may have to be collected, removed from the system, and disposed of in an acceptable manner. In the case of zinc, boron and fluorine the degree of volatilization has not yet been determined, but they would be expected to be rather volatile. Even if only 10% of the total amount is volatile, there will be large quantities to remove in the gas cleaning operation and to dispose of.

TABLE 4

TRACE ELEMENTS—ESTIMATED VOLATILITY

	Hypothetical Coal ppm	% Volatile*	lb/day**
Cl	1500	90+	54000
Hg	0.3	90+	10
Se	1.7	74	50
As	9.6	65	250
Pb	5.9	63	148
Cd	0.8	62	20
Sb	0.2	33	3
V	33	30	397
Ni	12	24	115
Be	0.9	18	7
Zn	44	e. g. 10	177
B	165	e. g. 10	660
F	85	e. g. 10	340
Ti	340	e. g. 10	1360
Cr	15	nil	nil

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* Volatility based mainly on gasification experiments (27) but chlorine is taken from combustion tests, while zinc, boron, and fluorine were taken at 10% for illustration in absence of data.

** Estimated volatility for 20,000 tons/day of coal to gasification.

A complication that has not generally been recognized, occurs in the gas cleanup section due to the volatility of trace elements. These are carried out with the raw gas, and will be removed in the gas cleanup facilities when the gas is cooled and scrubbed. In any event, they do not remain in the product gas, and it follows that they must leave the system at some point. Compounds such as cyanides might be destroyed by recycling to the process (e.g., the gasifier), but this can not be the case for elements such as arsenic, lead, chlorine, etc. Neither will they disappear in the biox unit. Therefore provision will be needed to separate and recover them, or to deactivate them for disposal in a satisfactory manner. As can be seen from Table 4, the combined amounts of all volatile portions of trace elements can present a formidable disposal problem.

The preceeding discussion has been directed primarily at trace elements that are partially volatilized during gasification and that therefore must be recovered and disposed of in the gas cleaning section. Consideration must also be given to trace metals that are not volatilized and leave in the solid effluents from the plant, one of which is the char from gasification. Undesirable elements might be leached out of this char if it is handled as a water slurry, and it will ultimately be exposed to leaching by ground water when it is disposed of as land fill or to the mine. Sufficient information is not now available to evaluate the potential problems and the situation may be quite different from the ash rejected from coal fired furnaces, since the char is produced in a reducing atmosphere rather than an oxidizing one. Background information on slag from blast furnaces used in the steel industry may be pertinent from this standpoint, since the blast furnace operates with a reducing atmosphere. However, a large amount of limestone is also added to the blast furnace, consequently the nature of the slag will be different.

9. TECHNOLOGY NEEDS

This review and examination of environmental aspects of the Winkler process has defined a number of areas where further information is needed to evaluate the situation, or where additional work could lead to significant improvement with regard to environmental impact, energy consumption, or thermal efficiency. Items of this nature will now be discussed, taken in the order of processing steps shown on the flowplan in Figure 1, and used in previous sections.

The first item to consider is coal drying. While thorough drying may not be needed or warranted, it is usually necessary to at least remove surface moisture in order to have reliable coal handling and feeding systems. Conventional dryers burn high value fuel and have a large volume of vent gas that must be cleaned up. An alternative to consider is using indirect heating, for example in a fluidized bed containing heating coils. Air might be recirculated through the bed and through condensers which would recover water that could be used as makeup. Heat might be supplied by low pressure steam if it is readily available from waste heat recovery. In other cases it may be possible to use waste heat that would otherwise be rejected to the atmosphere via air cooling. The advantage to be gained is that heat which must be rejected anyway is put to use. It also adds preheat to the coal feed, thereby decreasing heat load on the gasifier and oxygen consumption.

On gasification, if there were a way to make low purity oxygen at much lower energy consumption, the applications to provide clean fuel gas might then be more efficient. The oxygen plant is one of the largest consumers of utilities in the plant. Operating the gasifier at higher pressure will also save energy, particularly when the product gas is to be used at high pressure, as in a combined cycle. Even if the gas is burned in a low pressure furnace, an expander could be used to recover energy, if the gas is generated at high pressure. In other cases, the expander could be used to provide final cooling of the gas so as to save cooling water, or even to provide refrigeration.

As mentioned earlier, the char may be desulfurized during gasification to give a valuable low sulfur solid fuel. If so, it may be desirable to purposely maximize the yield of byproduct char. Techniques for augmenting desulfurization in the gasifier or by auxiliary facilities should be considered and evaluated as one approach. An alternative is to develop ways to obtain a high overall carbon conversion, so that the char contains little or no combustibles. Otherwise an effective way to recover the heating value in spent char is needed so as to avoid a large debit in thermal efficiency. One possibility is "clean combustion" in a fluid bed of limestone which serves as a sulfur acceptor.

On gas cleanup, a more effective way to remove dust would be useful. Even water scrubbing is not considered adequate in some commercial designs, and electrostatic precipitation is added. A dust removal system that can operate at elevated temperature would be desirable when using expanders or with a combined cycle application. Sand bed filters have been proposed for such service. A general discussion of alternatives for gas cleanup and sour water handling is given in reference 9.

The sulfur removal system often represents the largest single consumer of steam in a process for reboiling or stripping the solution used to absorb H_2S. A solvent having higher capacity may be helpful, possibly with operation at a higher pressure level. When making clean gas for fuel uses, it is not necessary to remove CO_2 and it is preferable to leave it in the gas when used in combined cycles. For such applications, more selective removal of sulfur would help and might save utilities in the regeneration step. Metals such as iron have been explored for desulfurization of gases, and should have the advantage of removing most forms of sulfur to a low level. These systems may be particularly useful when the operating pressure is low.

Cleanup of waste water for reuse consumes considerable energy, and is a difficult, complicated operation. Simpler, more effective and dependable systems would be useful. One possiblity is to use the adsorptive properties of the char, which would then be burned or circulated through the gasifier. A further discussion of considerations in waste water cleanup is given in reference 5.

Trace elements will also accumulate in the waste water. More information is needed on what happens to trace elements in the coal feed, where they appear, and in what form, so that satisfactory methods can be worked out for their recovery or disposal.

Water consumption by the plant is set largely by evaporation in the cooling tower. Therefore ways to minimize use of cooling water are of interest. Heat exchange and heat recovery should be maximized, while air cooling can then be used to decrease the amount of heat finally rejected to cooling water. In general, improvements in thermal efficiency and reduced utilities consumption will tend to save water. Practical ways to recover water from blowdown streams would also be desirable.

Additional discussion of technology needs will be found in earlier reports in this series.

10. PROCESS DETAILS

Further details on the basis used for this evaluation are given in Tables 5-10. A simplified flow diagram for the gasification section is shown in Figure 4.

TABLE 5

MAJOR INPUTS TO PLANT--WINKLER PROCESS

Coal to dryer	(13.3% moisture)	21,158 tons/day
Coal from dryer	(8.7% moisture)	20,100 tons/day

Coal Composition*	Wt %
Moisture	8.7
Carbon	54.1
Hydrogen	4.1
Oxygen	13.9
Nitrogen	0.6
Sulfur	3.3
Ash	15.3
	100.0

High heating value 9320 Btu/lb.

Plant makeup water - 14,362 tons/day

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* German dry brown coal. From reference 10, Table IV.

TABLE 6

MAJOR OUTPUTS FROM PLANT—WINKLER PROCESS

Net product gas (incl. 629 tons/day moisture)	22,920 tons/day (912 MM, scf)

Gas Composition (wet)	Vol. %
moisture	2.9
H_2	41.4
CO	37.8
CO_2	14.7
CH_4	1.9
N_2	1.2
H_2S + COS	0.1
	100.0

High heating value (wet) 274 Btu/scf

Char	
from gasifier (42% carbon)	924 tons/day
from cyclone (29% carbon)	3546 tons/day
Sulfur from sulfur plant	605 tons/day
Waste Water discharged from plant	1800 tons/day

Other: sludges and solids from treating waste and makeup water, dust from electrostatic precipitator, nitrogen (37,976 tons/day) from oxygen plant, plus gases from coal dryer, sulfur plant, and cooling tower.

TABLE 7

STEAM BALANCE--WINKLER PROCESS

	tons/day
600 psig steam	
Generated in gasifier	9045
Used in bleeder turbine exhausting at 35 psig. to supply all power needed in oxygen plant and to generate electricity for process. Exhaust steam at 35 psig. provides gasifier steam.	
125 psig steam	
Generated in waste heat boiler on raw gas	6030
Used in gasifier, acid gas removal, sour water stripping, etc.	

Note: plant is self-sufficient in utilities, so auxiliary steam and power generation are only needed for startup.

TABLE 8

ELECTRIC POWER REQUIRED--WINKLER PROCESS

	KW
Coal preparation	12,800
Gas scrubbing	600
Acid gas treatment	100
Gasifier	100
Sulfur plant	400
Cooling water pumps	3,000
Cooling tower fans	2,000
Oxygen plant and misc.	1,000
	20,000

This power is supplied by bleeder turbine on part of gasifier steam supply.

TABLE 9

WATER BALANCE--WINKLER PROCESS

	tons/day
Cooling Tower	
Evaporation	9,400
Drift loss	756
Blowdown	1,800
	11,956
From waste water treating	7,713
Fresh water makeup	4,243
Boiler Feed Water	
Steam to gasifier	9,849
Steam and condensate losses	270
Total BFW required	10,119
Fresh Water Makeup	
To cooling tower	4,243
To boiler feed water	10,119
	14,362
Net plant discharge of waste water (cooling tower blowdown)	1,800

Note: 3417 tons/day of treated sour water is used as quench at outlet of gasifier.

TABLE 10

MAKE UP CHEMICALS--WINKLER PROCESS

Chemicals

Acid Gas Removal:

- scrubbing solution
- additives

Sulfur Plant tail gas cleanup

Cooling Tower Additives

Anticorrosion, e. g. chromate
Antifouling, e. g. chlorine

Water Treating

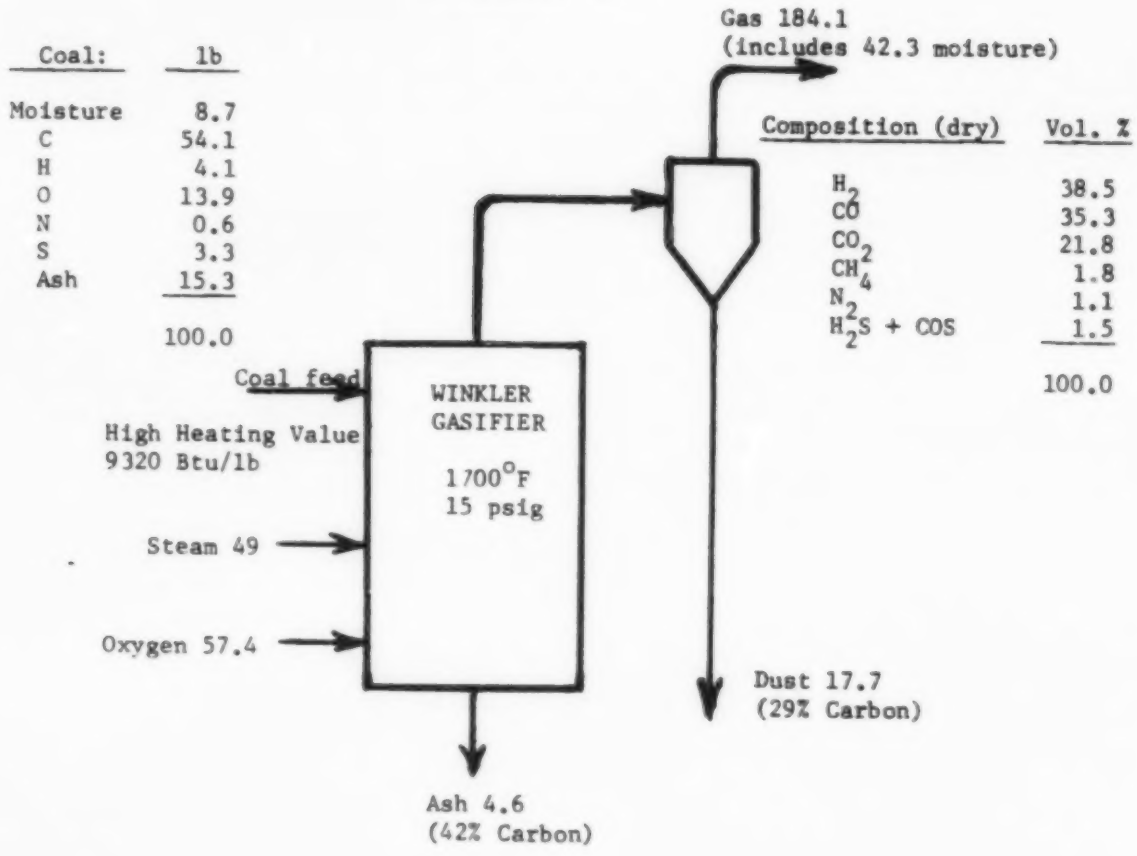
Lime
Alum
Caustic
Sulfuric Acid
Ion exchange resin

FIGURE 4

WINKLER GASIFIER USING OXYGEN

Numbers are pounds except as indicated

Reference: (10)



11. QUALIFICATIONS

As pointed out, this study does not consider cost or economics. Also, areas such as coal mining and general offsites are excluded, as well as miscellaneous small utility consumers such as instruments, lighting etc. These will be similar and common to all coal conversion operations.

The study is based on the specific process design and coal type cited, with modifications as discussed. Plant location is an important item of the basis and is not always specified in detail. It will affect items such as the air and water conditions available, and the type of pollution control needed. For example, this study uses high sulfur, lignite type coal for gasification. As mentioned earlier, the developer has indicated that oxygen consumption may be decreased in new plant designs. Because of variations in coal feed, moisture content, and other basic items, great caution is needed in making comparisons between coal gasification processes as they are not on a completely comparable basis.

The study is based on processing run of mine lignite. If bituminous coal were used, then coal cleaning would normally be needed with a considerable environmental impact as described in some other studies in this series (5). Refuse from coal cleaning may be 20-25% of the coal as mined, presenting a sizeable disposal problem.

Other gasification processes may make large amounts of various by-products such as tar, naphtha, phenols, and ammonia. The disposition and value of these must be taken into account relative to the increased coal consumption that results and the corresponding improvement in overall thermal efficiency. Such variability further increases the difficulty of making meaningful comparisons between processes.

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